

Spin-orbit coupling induced two-electron relaxation in silicon donor pairs

Yang Song^{1,2,*} and S. Das Sarma^{1,2}

¹*Condensed Matter Theory Center, Department of Physics,
University of Maryland, College Park, MD 20742, USA*

²*Joint Quantum Institute, University of Maryland, College Park, MD 20742, USA*

We unravel theoretically a key intrinsic relaxation mechanism among the low-lying singlet and triplet donor-pair states in silicon, an important element in the fast-developing field of spintronics and quantum computation. Despite the perceived weak spin-orbit coupling (SOC) in Si, we find that our discovered relaxation mechanism, combined with the electron-phonon and inter-donor interactions, dominantly drives the transitions in the two-electron states over a large range of donor coupling regime. The scaling of the relaxation rate with inter-donor exchange interaction J goes from J^5 to J^4 at the low to high temperature limits. Our analytical study draws on the symmetry analysis over combined band, donor envelope and valley configurations. It uncovers naturally the dependence on the donor-alignment direction and triplet spin orientation, and especially on the dominant SOC source from donor impurities. While a magnetic field is not necessary for this relaxation, unlike in the single-donor spin relaxation, we discuss the crossover behavior with increasing Zeeman energy in order to facilitate comparison with experiments.

PACS numbers:

A pair of coupled donors in an enriched Si²⁸ host crystal forms the most cleanly defined two-qubit system or singlet-triplet (S - T) qubit in solid state materials [1, 2]. The nearly noise-free crystal environment [3–5] together with the reproducible donor properties fixed by nature [6] places an essential role for the intrinsic energy relaxation among the lowest two-electron states by electron-phonon (e-ph) interaction in studying the coherence limit of quantum computation involving such qubits [7, 8]. A proper treatment of this problem with a transparent and physical basis is imperative, enabling its various modifications such as proximity with interface [9], gate potential [10, 11], or quantum dots [12, 13] which are necessary for qubit control and operation.

Electron spin relaxation of single donor states in Si has been studied since 1950s. Much interest then was associated with the ensemble spin resonance experiments which could remarkably map out the detailed donor wavefunction by its hyperfine interaction with donor or Si²⁹ nucleus [14, 15]. Theoretical attention was thus originally paid to the simultaneous flip of electron and donor nucleus spins (T_x) [16] through hyperfine interaction, until later the importance of spin-orbit coupling (SOC) driven spin relaxation was established [17–20]. However, the two-electron relaxation on a coupled donor pair has been much less studied over the decades, with only a few in the context of concentration-dependent spin relaxation [21] or of quantum computation [7]. Both of these previous studies focused on the hyperfine interaction to mix S and T states, while neglecting SOC as too weak in Si. In this Letter, we study the donor-pair spin relaxation in Si using fundamental symmetry properties.

Here we unravel a new *general* relaxation mechanism among two-electron states to address this important long-standing problem in the context of Si donor-based spin qubits. This robust mechanism has a parametrically

dominating J^5 dependence on the 2-donor exchange coupling J for the $S \leftrightarrow T$ transition, and is crucial except for the very weak coupling regime (which is of little interest in the context of quantum computation). This mechanism relies only on the intrinsic SOC in the system to couple spins, with the donor SOC dominating the Si host SOC, but not on any structure-induced Rashba or Dresselhaus field. This newly-found dominance of the donor SOC is absent in the classical single-donor spin relaxation by g -factor modulation [17]. The effectiveness of the two SOC, i.e., of the host and the impurity, is considered by taking full account of the three physical ‘layers’ in the problem: the Bloch bands, the hydrogenic-like donor envelopes, and the multi-valley configurations. Within each layer rigorous selection rules are enforced for e-ph, SOC and interdonor perturbations, while at the same time, the decoupling between the underlying fast-oscillatory physics and the slowly-varying localized envelopes realizes an efficient leading-order quantitative estimation. The unique donor and spin alignment dependence naturally falls out of our analytical treatment. Finally, we elucidate the crossover from low to high Zeeman energy as a reduction of the two-electron problem into a single-electron one, although the presence of spin splitting is not a priori essential for our novel mechanism.

We start with a set of two-electron representations as general as necessary satisfying the symmetry of the specific two-donor system. While these states mainly comprise the Heitler-London states out of unperturbed single-donor ground states, both nonpolar and polar (‘ionic’) mixtures are included. These excited components are needed for nonzero e-ph and SOC coupling, and are mixed in by the interdonor interaction. The dominant effect comes from the leading-order mixture where one of the two electrons occupies the single-donor ground state. As a result, the general states follow the concise

expressions,

$$S : \frac{1 - \mathcal{P}_{12}}{2\sqrt{1 + \chi^2}} [\psi_{\alpha\uparrow}^S(\mathbf{r}_1)\psi_{\beta\downarrow}^S(\mathbf{r}_2) - \psi_{\alpha\downarrow}^S(\mathbf{r}_1)\psi_{\beta\uparrow}^S(\mathbf{r}_2)], \quad (1)$$

$$T_0 : \frac{1 - \mathcal{P}_{12}}{2\sqrt{1 - \chi^2}} [\psi_{\alpha\uparrow}^{T_0}(\mathbf{r}_1)\psi_{\beta\downarrow}^{T_0}(\mathbf{r}_2) + \psi_{\alpha\downarrow}^{T_0}(\mathbf{r}_1)\psi_{\beta\uparrow}^{T_0}(\mathbf{r}_2)], \quad (2)$$

$$T_{\pm} : \frac{1 - \mathcal{P}_{12}}{\sqrt{2(1 - \chi^2)}} [\psi_{\alpha\uparrow(\downarrow)}^{T_{\pm}^{(-)}}(\mathbf{r}_1)\psi_{\beta\uparrow(\downarrow)}^{T_{\pm}^{(-)}}(\mathbf{r}_2)], \quad (3)$$

where \mathcal{P}_{12} exchanges electrons 1 and 2, $\alpha(\beta)$ labels the donor around which a one-electron wavefunction $\psi_{\alpha(\beta)}$ is *mainly* located, $\chi = \langle \psi_{\alpha} | \psi_{\beta} \rangle$, superscripts ($S, T_{0,\pm}$) distinguish different mixture components, and finally spin \uparrow and \downarrow is along the $\alpha - \beta$ donor-alignment direction ($\hat{\mathbf{d}}$), dictated by symmetry.

We next classify the two-electron states and the e-ph interaction by the all-relevant system symmetries for three crystallographic $\hat{\mathbf{d}}$, listed in Table I. For brevity, we use strain element ϵ_{ij} in place of the e-ph interaction $\sim [r_i dV/dr_j + (i \leftrightarrow j)]/2$ which transform the same symmetry-wise. For $\hat{\mathbf{d}} \parallel [110]$, S or $T_- + T_+$ denotes the eigenstate with the *majority* S or $T_- + T_+$ component respectively since they belong to the same irreducible representation (IR).

TABLE I: For each $\hat{\mathbf{d}}$, [001], [111] or [110], we find its point group and the IRs of the states and phonon modes. For $\hat{\mathbf{d}} \parallel [111]$, two subcases, with inversion symmetry (i) and without it (n), depend on the alternating donor positions (even though individual donors possess no inversion symmetry). To be definite, we set $z' \parallel [111]$, $x' \parallel [10 - 1]$, $y' \parallel [-12 - 1]$, $z'' \parallel [110]$, $x'' \parallel [001]$ and $y'' \parallel [1 - 10]$.

	[001] (D_{2d})	[111] n (C_{3v}) [111] i (D_{3d})	[110] (C_{2v})
S	A_1	A_1	A_1
T_0	B_1	$A_2(n), A'_1(i)$	A_2
T_{\pm}	E	$E(n), E'(i)$	$A_1(T_+ + T_-)$ $B_2(T_+ - T_-)$
	$A_1 \begin{pmatrix} \epsilon_{zz} \\ \epsilon_{xx} + \epsilon_{yy} \end{pmatrix}$	$A_1 \begin{pmatrix} \epsilon_{z'z'} \\ \epsilon_{x'x'} + \epsilon_{y'y'} \end{pmatrix}$	$A_1 \begin{pmatrix} \epsilon_{x''x''} \\ \epsilon_{y''y''} \end{pmatrix}$
ϵ_{ij}	$B_1 \begin{pmatrix} \epsilon_{xx} - \epsilon_{yy} \\ B_2(\epsilon_{xy}) \\ E(\{\epsilon_{xz}, \epsilon_{yz}\}) \end{pmatrix}$	$E \begin{pmatrix} \{\epsilon_{x'x'} - \epsilon_{y'y'}\} \\ 2\epsilon_{x'y'} \\ \{\epsilon_{x'z'}, \epsilon_{y'z'}\} \end{pmatrix}$	$B_1 \begin{pmatrix} \epsilon_{x''x''} \\ B_2(\epsilon_{x''y''}) \\ A_2(\epsilon_{y''z''}) \end{pmatrix}$

This top-down symmetry approach immediately identifies the allowed relaxation transitions and the associated specific acoustic phonon modes (e.g., $\langle T_0 | \epsilon_{xx} - \epsilon_{yy} | S \rangle_{[001]}$), independent of the quantitative treatments one adopts. We also take into account the fact that S and T states (e-ph operator) are two(one)-body objects, removing all $\langle T_- | \epsilon_{ij} | T_+ \rangle$ where $\langle \psi_{\alpha(\beta)\downarrow}^{T_-} | \psi_{\alpha\uparrow}^{T_+} \rangle$ are always strictly zero, solely by the $C_{2(3)}$ operation in the $\hat{\mathbf{d}} \parallel [001]$ ([111]) case.

To evaluate the allowed relaxation matrix elements, the first rounds of reduction come by substituting the forms of S and T states with Eqs. (1)-(3). Even though

the specific ψ 's are altered away from the single-donor wavefunction by interdonor interaction, they obey precise symmetry relations among themselves due to the point group of a given two-donor system. Utilizing these relations [22], the matrix elements are reduced into products of single-electron ones,

$$\langle T_0 | \epsilon_{xx} - \epsilon_{yy} | S \rangle_{[001]} \quad (4)$$

$$\propto \sum_{\gamma=\alpha,\beta} (-1)^{\gamma} \langle \psi_{\gamma\uparrow}^{T_0} | \epsilon_{xx} - \epsilon_{yy} | \psi_{\alpha\uparrow}^S \rangle \langle \psi_{\gamma\downarrow}^{T_0} | \psi_{\beta\downarrow}^S \rangle, \quad (5)$$

$$\langle T_0 | \epsilon_{y''z''} | S \rangle_{[110]} \quad (6)$$

$$\propto \sum_{\substack{\uparrow=\uparrow,\downarrow \\ \gamma=\alpha,\beta}} (-1)^{\gamma} \langle \psi_{\gamma\uparrow}^{T_0} | \epsilon_{y''z''} | \psi_{\alpha\uparrow}^S \rangle \langle \psi_{\gamma\downarrow}^{T_0} | \psi_{\beta\downarrow}^S \rangle, \quad (7)$$

$$\langle T_+ | \epsilon_{xz} \mp i\epsilon_{yz} | S/T_0 \rangle_{[001]} \quad (8)$$

$$\propto \frac{\mp 1}{\sqrt{2}} \sum_{\gamma=\alpha,\beta} (-1)^{\gamma} \langle \psi_{\gamma\uparrow}^{T_+} | \epsilon_{xz} \mp i\epsilon_{yz} | \psi_{\alpha\downarrow}^{S/T_0} \rangle \langle \psi_{\gamma\uparrow}^{T_+} | \psi_{\beta\uparrow}^{S/T_0} \rangle, \quad (9)$$

$$\langle T_+ | \epsilon_{x'z'} + i\epsilon_{y'z'} | S/T_0 \rangle_{[111]n} \quad (10)$$

$$\propto \frac{\mp 1}{2\sqrt{2}} \sum_{\gamma,\gamma'=\alpha,\beta} (-1)^{\gamma+\gamma'+\frac{\gamma'}{2}} \langle \psi_{\gamma\uparrow}^{T_+} | \epsilon_{x'z'} + i\epsilon_{y'z'} | \psi_{\alpha\downarrow'}^{S/T_0} \rangle \langle \psi_{\gamma\uparrow'}^{T_+} | \psi_{\beta\uparrow'}^{S/T_0} \rangle, \quad (11)$$

$$\langle T_+ | \epsilon_{x'z'} + i\epsilon_{y'z'} | T_0 \rangle_{[111]i} \quad (12)$$

$$\propto \frac{1}{\sqrt{2}} \sum_{\gamma=\alpha,\beta} (-1)^{\gamma} \langle \psi_{\gamma\uparrow}^{T_+} | \epsilon_{x'z'} + i\epsilon_{y'z'} | \psi_{\alpha\downarrow'}^{T_0} \rangle \langle \psi_{\gamma\uparrow}^{T_+} | \psi_{\beta\uparrow}^{T_0} \rangle, \quad (13)$$

$$\langle T_+ \pm T_- | \epsilon_{A_1} / \epsilon_{x''y''} | S \rangle_{[110]} \quad (14)$$

$$\propto \sum_{\substack{\uparrow=\uparrow,\downarrow \\ \gamma=\alpha,\beta}} (-1)^{\uparrow+\gamma} \langle \psi_{\gamma\uparrow}^{T_{1/2}} | \epsilon_{A_1/x''y''} | \psi_{\alpha\uparrow}^S \rangle \langle \psi_{\gamma\uparrow}^{T_{1/2}} | \psi_{\beta\uparrow}^S \rangle, \quad (15)$$

$$\langle T_+ \pm T_- | \epsilon_{y''z''} / \epsilon_{x''z''} | T_0 \rangle_{[110]} \quad (16)$$

$$\propto \sum_{\substack{\uparrow=\uparrow,\downarrow \\ \gamma=\alpha,\beta}} (-1)^{\gamma} \langle \psi_{\gamma\uparrow}^{T_{1/2}} | \epsilon_{y''z''} / \epsilon_{x''z''} | \psi_{\alpha\downarrow}^{T_0} \rangle \langle \psi_{\gamma\uparrow}^{T_{1/2}} | \psi_{\beta\downarrow}^{T_0} \rangle, \quad (17)$$

$$\langle T_+ - T_- | \epsilon_{x''y''} | T_+ + T_- \rangle_{[110]} \quad (18)$$

$$\propto \sum_{\substack{\uparrow=\uparrow,\downarrow \\ \gamma=\alpha,\beta}} (-1)^{\gamma+\uparrow} \langle \psi_{\gamma\uparrow}^{T_2} | \epsilon_{x''y''} | \psi_{\alpha\uparrow}^{T_1} \rangle \langle \psi_{\gamma\uparrow}^{T_2} | \psi_{\beta\uparrow}^{T_1} \rangle, \quad (19)$$

where for brevity we use ' \propto ' and omit the normalization factor $1/\sqrt{1 \pm \chi^2}$, $\bar{\gamma}$ or $\bar{\uparrow}$ denotes the opposite donor or spin respectively, at exponent $\alpha, \uparrow \equiv 0$ and $\beta, \downarrow \equiv 1$, \uparrow' (\uparrow'') marks that the spin is along the z' (z'') rather than z direction, and $T_{1/2} \equiv T_+ \pm T_-$ in Eqs. (10) and (11). The transition magnitudes from S or T_0 to T_- state are the same as those to T_+ by switching between $\epsilon_{xz} \pm i\epsilon_{yz}$ ($\hat{\mathbf{d}} \parallel [001]$) or $\epsilon_{x'z'} \pm i\epsilon_{y'z'}$ ($\hat{\mathbf{d}} \parallel [111]$). In $\hat{\mathbf{d}} \parallel [111]$, one can just substitute $\epsilon_{x'z'} \pm i\epsilon_{y'z'}$ with $\epsilon_{x'x'} - \epsilon_{y'y'} \pm 2i\epsilon_{x'y'}$. We note that the additional inversion in the [111] i case equates each of the two pairs in Eq. (7) of [111] n , nullifying $\langle T_{\pm} | \epsilon_{x'z'} \pm i\epsilon_{y'z'} | S \rangle_{[111]i}$ while leading to Eq.(8). This set of equations arising from very general symmetry considerations constitutes one of the key results in this work.

We apply perturbation theory to quantify the single-electron matrix elements in terms of SOC and exchange

coupling constants, in addition to the deformation potential coupling. The first two perturbations are necessary, as without them the transitions reduce to ones between pure opposite spins or single-donor spin relaxation which are known to vanish. In particular, the same-spin e-ph matrix element in Eq. (4) from $\langle T_0 | \epsilon_{xx} - \epsilon_{yy} | S \rangle_{[001]}$ requires a z -component SOC operator, since

$$\langle \psi_{\alpha(\beta)\uparrow}^{T_0} | \epsilon_{xx} - \epsilon_{yy} | \psi_{\alpha\uparrow}^S \rangle = -\langle \psi_{\alpha(\beta)\downarrow}^{T_0} | \epsilon_{xx} - \epsilon_{yy} | \psi_{\alpha\downarrow}^S \rangle \quad (12)$$

by the $\sigma_{[110]}$ reflection symmetry. It is similar for the same-spin transitions of $\hat{\mathbf{d}} \parallel [110]$ in Eqs. (5), (10) and (11) except with the ϵ_{A_1} or $\epsilon_{x''z''}$ modes, while all the rest manifestly require SOC to flip spin. To proceed within the perturbation theory, we choose our basis states to be the spinless donor states,

$$\{\psi_k\} = \sum_{i=1}^6 v_{k,i} F_{k,i}^{nlm}(\mathbf{r}) \psi_{k,i}^{\Delta_j}, \quad (13)$$

which are identified by three indices: the Si bulk band ($\Delta_1, \Delta_2, \Delta_3, \Delta_4$ and Δ_5) [23], the donor envelope with an orbital number nlm (with ellipsoidal effective mass) [24], and the T_d (tetrahedral) group IR (A_1, A_2, E, T_1 and T_2) [25] which determines $v_{k,i}$ ($\sum_i |v_{k,i}|^2 = 1$) considering the participating Δ_j and nlm . These three indices fix the energy level in a roughly descending order. Before exhaustively working out all possible selection rules among this multitude of states, we examine the essential physics of coupling strengths for different perturbation interactions and select the stronger coupling efficiently.

First, we focus on the interdonor interaction, including direct Coulomb and exchange. It couples donor states made of different bulk Δ bands very weakly. As we know, in single donor ground states, bands other than the conduction Δ_1 band are routinely neglected due to their fast-oscillating difference and the slow-varying nature of the Coulomb interaction [6, 26]. Here the coupling by interdonor interaction is even weaker as the donor distance is several times the Bohr radius. Within the same bulk band, it can couple different donor envelopes as well as valley configurations effectively, as their differences are (partly) slowly varying. Second, the e-ph coupling between the same or different Δ_j 's are efficient when allowed by symmetry, as the interaction involves periodic ion potentials. This gives rise to various intraband and interband deformation potentials. Once the interaction matches the symmetry difference of the two bulk bands, it can only couple the same envelopes as no extra symmetry from the phonon mode compensates for the different envelope symmetries. However, it may couple different valley configurations as the intravalley e-ph coupling may change from valley to valley. Last, we discuss the SOC of

two different types arising from the host and the donor [27]. For Si host SOC, it couples symmetry-allowed Δ bands strongly but different envelopes negligibly just like the e-ph coupling. The donor impurity SOC may couple donor envelopes in the same band effectively [20, 31, 32]. Additionally, both SOC's can couple different valley configurations allowed by symmetry. However, the exclusive intravalley coupling nature of the host SOC together with that of e-ph coupling suppresses their effect by a very large factor of the relevant phonon wavelength divided by lattice constant (i.e., the ‘‘Elliott-Yafet’’ cancellation [33]).

Understanding the potentially dominant couplings, one can locate the symmetry-allowed ones among them. The e-ph and SOC couplings follow conventional single-particle selection rules, as discussed below. The two-body interdonor interaction, however, requires a different treatment. We identify the allowed T_d IRs for the mixed single-donor states, such that under every two-donor symmetry operation the resulting $\psi_{\gamma\uparrow}$ transforms the same as that for the ground state [22]. For instance, $\psi_{\alpha\uparrow}$ in Eqs. (1)-(3) for $\hat{\mathbf{d}} \parallel [001]$ follows,

$$\begin{aligned} \alpha\uparrow = & \alpha_{A_1\uparrow} + \delta_0^\beta \beta_{A_1\uparrow} + \sum_{\gamma=\alpha,\beta} (i\delta_1^\gamma \gamma_{A_2\uparrow} \\ & + i\delta_2^\gamma \gamma_{E_z^I\uparrow} + \delta_3^\gamma \gamma_{E_z^H\uparrow} + \delta_4^\gamma \gamma_{T_{1z}\uparrow} + i\delta_5^\gamma \gamma_{T_{2z}\uparrow}), \end{aligned} \quad (14)$$

where ‘ ψ ’ is omitted for shortness, and both the nonpolar ($\gamma = \alpha$) and polar (β) mixtures are included with time-reversal (TR) compatible phases and small real coefficients δ_i^γ 's. Each δ is distinct for different states except in the same IR (e.g., T_\pm of [001] in Table I). Without crystal anisotropy, its difference between S and T is due to the exchange part (as opposed to the direct Coulomb), and $\delta_0^\beta = 0$ for T states due to Pauli exclusion.

The perturbation theory for relaxation matrix elements throughout Eqs. (4)-(11) then proceeds in a straightforward manner following the above preparations [22]. We find the symmetries of involved basis states and interaction operators, and evaluate various perturbation integrals. Here we illustrate the key common aspects by analyzing the representative Eq. (4) in more detail. Both terms require interdonor interaction amounting to two overlap factors, so neither of them may be neglected a priori. As shown in Eq. (12), SOC is necessary. We find that it is dominated by donor SOC ($\sim \lambda_{soc} \mathbf{L} \cdot \mathbf{s}$). The donor L_z and $\epsilon_{xx} - \epsilon_{yy}$ bring Δ_1 band back to itself allowing the remaining interdonor coupling (H_{int-d}). $\epsilon_{xx} - \epsilon_{yy}$, moreover, connects two available valley configurations comprising Δ_1 -1s states. Together, the $\gamma = \alpha$ term in Eq. (4) contains a perturbation expansion,

$$\langle T_0 | \epsilon_{xx} - \epsilon_{yy} | S \rangle_{[001]}^{(1)} \propto \sum_{\nu=1s, 3d_{\pm 1}} \frac{\langle \alpha_{\Delta_1, 1s, A_1}^{T_0} | H_{int-d} | \alpha_{\Delta_1, \nu, T_{2z}}^{T_0} \rangle \langle \alpha_{\Delta_1, \nu, T_{2z}} | L_z | \alpha_{\Delta_1, 1s, E_z^I} \rangle \langle \alpha_{\Delta_1, 1s, E_z^I} | \epsilon_{xx} - \epsilon_{yy} | \alpha_{\Delta_1, 1s, A_1} \rangle}{(\mathcal{E}_{\Delta_1, 1s, A_1} - \mathcal{E}_{\Delta_1, \nu, T_2})(\mathcal{E}_{\Delta_1, 1s, A_1} - \mathcal{E}_{\Delta_1, 1s, E})}, \quad (15)$$

plus another one with reversed ordering of interactions, where H_{int-d} couples A_1 and T_{2z} envelopes in the same donor [as expected from Eq. (14)] of the S instead of T_0 state. That leaves only the exchange part of H_{int-d} effective ($\sim J_{A_1 T_2}$). $\mathcal{E}_{\Delta_1, 1s, A_1} - \mathcal{E}_{\Delta_1, 1s, E/T_2}(\mathcal{E}_{\Delta_1, 3d_{\pm 1}, T_2}) \approx -12(40)$ meV [36], and $\langle E_z^I | \epsilon_{xx} - \epsilon_{yy} | A_1 \rangle = \sqrt{2/3} \Xi_u$ with $\Xi_u \approx 8.77$ eV [37]. Two major donor SOC couplings emerge: one is between two $1s$ configurations and relates to the donor spin split $\Delta_{SOC}^{dnr} (\sim 0.03, 0.1, 0.3$ meV for P, As, Sb donors respectively) [20, 28], and the other is between $1s$ and $3d_{\pm 1}$ where $\langle 3d_{\pm 1} | L_z | 1s \rangle \neq 0$ within a *single* (x or y) valley due to anisotropy of the $1s$ envelope [24]. We find the $\nu = 1s$ component can be safely used for an order of magnitude estimate [38]. For the $\gamma = \beta$ term in Eq. (4), the $J_{A_1 T_2}$ factor is replaced by an exchange term with the polar state $\beta_{A_1} \beta_{T_2}$ multiplied by another overlap factor, hence the same order of magnitude. By re-ordering the interactions, we see that two other perturbation expansions of the similar or smaller magnitude are allowed (see [22] for more details).

More perturbation terms of similar magnitudes exist, representing the combined interaction operators. The typical example is the *Yafet* term in the Elliott-Yafet spin flip mechanism [33]. Others include phonon-modulated exchange, exchange SOC, etc. It is not practical to enumerate each term, however. Even in the much simpler pure bulk Si, numerous comparable leading-order terms contribute [30]. In the single-donor relaxation, for example, Yafet term is not considered [17]. More importantly, it is neither essential to do so since these terms possess the same dependence on operator components (e.g., ϵ_{ij}, s_i) according to the method of invariants [34, 35], justifying this treatment. The net numerical prefactors are difficult to evaluate exactly, and should be left to be extracted experimentally aided by our transparent expressions.

Following the analysis, we obtain the leading-order magnitude $|M|$ for relaxation channels $\langle T_0 | \dots | S \rangle_{[110]}$, $\langle T_{\pm} | \dots | S \rangle_{[111]n}$, and $\langle T_+ + T_- | \dots | S \rangle_{[110]}$ (indexed by $\kappa = 2, 3, 4$ whereas $\langle T_0 | \dots | S \rangle_{[001]}$ by $\kappa = 1$) via donor SOC,

$$M_{\kappa, \lambda} \approx \epsilon_{\kappa, \lambda}^{ph} \mathcal{F}_{\kappa} \frac{J_{A_1 T_2} \cdot \sqrt{\frac{2}{3}} \Xi_u \cdot \Delta_{SOC}^{dnr}}{\Delta \mathcal{E}_1 \Delta \mathcal{E}_2}, \quad (16)$$

where λ is the phonon mode, $\Delta \mathcal{E}_{1(2)} = -12$ meV, channel-dependent factor $\mathcal{F}_{1,2,3,4} = 1, \frac{1}{\sqrt{2}}, \frac{3\sqrt{2}i}{8}, \frac{1}{\sqrt{2}}$, respectively, and $\epsilon_{\kappa, \lambda}^{ph} = \tilde{\epsilon}_{\kappa, \lambda} \sqrt{\hbar(n_{q\lambda} + 1)/2\rho\omega_{\lambda}(q)}$. $\tilde{\epsilon}_{\kappa, \lambda}$ for $\kappa = 1, 2$ is $\frac{1}{2}(\tilde{\epsilon}_{xx} - \tilde{\epsilon}_{yy})$, for $\kappa = 3$ is $\frac{1}{3\sqrt{2}}(e^{i\frac{\pi}{6}}\tilde{\epsilon}_{xx} -$

$i\tilde{\epsilon}_{yy} - e^{-i\frac{\pi}{6}}\tilde{\epsilon}_{zz} + e^{-i\frac{\pi}{6}}\tilde{\epsilon}_{xy} + i\tilde{\epsilon}_{xz} - e^{i\frac{\pi}{6}}\tilde{\epsilon}_{yz})$ and for $\kappa = 4$ is $\tilde{\epsilon}_{zz} + \frac{1}{2}(\tilde{\epsilon}_{xx} + \tilde{\epsilon}_{yy})$ where $\tilde{\epsilon}_{ij} \equiv i(q_i \xi_j + q_j \xi_i)/2$, and then phonon polarization $\boldsymbol{\xi}(\mathbf{q})$ is projected into $\lambda = \text{LA, TA}_1, \text{TA}_2$ by elastic continuum approximation [40]. $\rho = 2.33$ g/cm³, $\hbar\omega_{\lambda} = \hbar v_{\lambda} q$ equals the relaxation energy, $v_{\text{LA(TA)}} = 8.7(5) \times 10^5$ cm/s, and n_q is the phonon distribution. Eq. (16) is not expected to be exact for the reason discussed above. Our goal is to provide the leading-order estimate for M and its robust dependence on J and temperature. We find for $\langle T_{\pm} | \dots | S \rangle_{[001]}$ and $\langle T_+ - T_- | \dots | S \rangle_{[110]}$ the leading-order term vanishes by symmetry and effective-mass analysis [22] and our mechanism is overshadowed by hyperfine-induced relaxation [7]. M between T states are also obtained with a critical difference regarding the interdonor coupling. Unlike between S and T states, its effect stems from the anisotropic SOC part of H_{int-d} and scales down roughly by a large factor $|J/I|$ where $I = \mathcal{E}_{T_0} - \mathcal{E}_{T_{\pm}}$.

Our result differs from the existing hyperfine-coupling theory on two-donor relaxation in Refs. [21] and [7] in a critical way that the spin mixing scales with $|\text{SOC coupling/spinless coupling}| (\sim 0.03 \text{ meV}/12 \text{ meV})$ instead of $|\text{hyperfine}(A)/\text{exchange}(J)|$ ($A \sim 0.2 \mu\text{eV}$ and J is on the order of 1 meV for $d \sim 8$ nm). Our mechanism is stronger than the hyperfine mechanism for larger exchange coupling as necessary for qubit operations. Also our stronger scaling on J (by J^2) offers a clear experimental distinction between the two mechanisms.

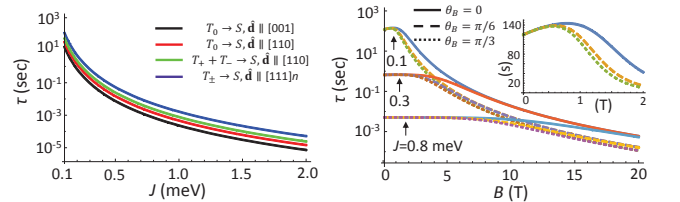


FIG. 1: (a) Relaxation time τ for four leading-order channels driven by donor SOC as $0.1 \text{ meV} < J < 2 \text{ meV}$. (b) τ of $T_+ \rightarrow S$ and $\hat{d} \parallel [111]n$ for three different J 's as $B < 20$ T. The inset shows an increasing τ at $g\mu_B B \lesssim k_B T$, a feature of finite exchange coupling. $T = 1\text{K}$ and the single-electron relaxation uses experimental parameters from [18].

Figure 1(a) quantifies the relaxation time τ for exchange coupling between 0.1 and 2 meV, by $\tau^{-1} = \frac{2\pi}{\hbar} \int \frac{d^3 q}{(2\pi)^3} \sum_{\lambda} |M_{\lambda}|^2 \delta(J - \hbar v_{\lambda} q_{\lambda})$ via Eq. (16) and $\int d\Omega_{\mathbf{q}} |\tilde{\epsilon}_{\kappa, \lambda}|^2$ [41]. $\tau \propto J^{-5}$ ($k_B T \ll J$) or TJ^4 ($k_B T \gg J$). The dependence on the phonon mode, which is rigorous from Eqs. (4)-(11), is made explicit. The nu-

merical factor due to interdonor interaction, however, is crudely averaged as exchange splitting J . Various exchange terms from different perturbation expansions [as discussed between Eqs. (15) and (16)] lead to superposed oscillations over donor vector \mathbf{d} , unrealistic for a perturbation calculation like ours to specify.

Finally, TR forbids spin relaxation by SOC on a single donor strictly at zero magnetic (B) field [17] whereas it does not affect any transition among the lowest two-electron states. As a result, the application of B field has most effect on driving one-electron spin flips, which dominate electron relaxation when $g\mu_B B \gtrsim J$. Indeed, we see a crossover behavior with increasing B in Fig. 1(b) for an $S \rightarrow T_-$ relaxation. The crossover is much sooner for $T_0 \rightarrow T_-$ as $g\mu_B B \sim I$. In fact, since the B power-law dependence for single donors and our J or I dependence are exactly the same for any temperature, the crossover occurs at a fixed $g\mu_B B/J(I)$ ratio depending only on the donor type.

In conclusion, we have established using rigorous symmetry considerations that, contrary to popular belief, spin-orbit coupling could produce a strong 2-electron relaxation in coupled donor qubits in Si in spite of vanishing Rashba and Dresselhaus effects.

This work is supported by LPS-MPO-CMTC.

* Electronic address: ysong128@umd.edu

- [1] F. A. Zwanenburg, A. S. Dzurak, A. Morello, M. Y. Simmons, L. C. L. Hollenberg, G. Klimeck, S. Rogge, S. N. Coppersmith, and M. A. Eriksson, *Rev. Mod. Phys.* **85**, 961 (2013).
- [2] S. Shankar, A. M. Tyryshkin, and S. A. Lyon, *Phys. Rev. B* **91**, 245206 (2015).
- [3] A. M. Tyryshkin, S. A. Lyon, A. V. Astashkin, and A. M. Raitsimring, *Phys. Rev. B* **68**, 193207 (2003).
- [4] J. T. Muhonen, J. P. Dehollain, A. Laucht, F. E. Hudson, R. Kalra, T. Sekiguchi, K. M. Itoh, D. N. Jamieson, J. C. McCallum, A. S. Dzurak, and A. Morello, *Nat. Nanotech.* **9**, 986 (2014).
- [5] K. M. Itoh and H. Watanabe, *MRS Commun.* **4**, 143 (2014).
- [6] W. Kohn, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1957), Vol. 5.
- [7] M. Borhani and X. Hu, *Phys. Rev. B* **82**, 241302(R) (2010).
- [8] Y.-L. Hsueh, H. Büch, Y. Tan, Y. Wang, L. C. L. Hollenberg, G. Klimeck, M. Y. Simmons, and R. Rahman, *Phys. Rev. Lett.* **113**, 246406 (2014).
- [9] M. J. Calderón, B. Koiller, and S. Das Sarma, *Phys. Rev. B* **75**, 125311 (2007).
- [10] J. P. Dehollain, J. T. Muhonen, K. Y. Tan, A. Saraiva, and D. N. Jamieson, *Phys. Rev. Lett.* **112**, 236801 (2014).
- [11] M. F. Gonzalez-Zalba, A. Saraiva, M. J. Calderón, D. Heiss, B. Koiller, and A. J. Ferguson, *Nano Lett.* **14**, 5672 (2014).
- [12] R. H. Foote, D. R. Ward, J. R. Prance, J. K. Gamble, E. Nielsen, B. Thorgrimsson, D. E. Savage, A. L. Saraiva, M. Friesen, S. N. Coppersmith, and M. A. Eriksson, *Appl. Phys. Lett.* **107**, 103112 (2015).
- [13] P. Harvey-Collard, N. T. Jacobson, M. Rudolph, J. Dominguez, G. A. T. Eyck, J. R. Wendt, T. Pluym, J. K. Gamble, M. P. Lilly, M. Pioro-Ladrière, and M. S. Carroll, arXiv:1512.01606.
- [14] R. C. Fletcher, W. A. Yager, G. L. Pearson, A. N. Holden, W. T. Read, and F. R. Merritt, *Phys. Rev.* **94**, 1392 (1954).
- [15] G. Feher, *Phys. Rev.* **114**, 1219 (1959).
- [16] D. Pines, J. Bardeen, and C. P. Slichter, *Phys. Rev.* **106**, 489 (1957).
- [17] H. Hasegawa, *Phys. Rev.* **118**, 1523 (1960); L. M. Roth, *Phys. Rev.* **118**, 1534 (1960); L. Roth, Massachusetts Institute of Technology Lincoln Laboratory Reports, April, 1960.
- [18] D. K. Wilson and G. Feher, *Phys. Rev.* **124**, 1068 (1961).
- [19] T. G. Castner, *Phys. Rev.* **130**, 58 (1963).
- [20] T. G. Castner, *Phys. Rev.* **155**, 816 (1967).
- [21] K. Sugihara, *J. Phys. Chem. Solids* **29**, 1099 (1968).
- [22] Supplemental materials where we provide explicit symmetry operation tables and the derivation details for Eqs. (4)-(11), allowed mixed IRs and expressions for δ in Eq. (14) and other cases, estimation for major perturbation expansions besides Eq. (15).
- [23] M. Lax and J. J. Hopfield, *Phys. Rev.* **124**, 115 (1961).
- [24] W. Kohn and J. M. Luttinger, *Phys. Rev.* **98**, 915 (1955).
- [25] C. J. Bradley and A. P. Cracknell, p433, *The Mathematical Theory of Symmetry in Solids: Representation Theory for Point Groups and Space Groups*, (Clarendon Press, Oxford, 1972).
- [26] J. M. Luttinger and W. Kohn, *Phys. Rev.* **97**, 869 (1955).
- [27] The donor SOC was not considered, for example, in Refs. [17] due to the measured small dependence of electron g -factor on donor types [15] indicating its weak interband coupling. On the other hand, the strong donor-dependent spin split clearly manifests the relevance of donor SOC [20, 28], which leads to comparable or stronger spin mixing (~ 0.03 meV/12 meV for Si:P) than that induced by host SOC (~ 4.1 meV/4.3 eV, see, e.g., [29, 30]).
- [28] Y. Song, O. Chalaev, and H. Dery, *Phys. Rev. Lett.* **113**, 167201 (2014).
- [29] P. Li and H. Dery, *Phys. Rev. Lett.* **107**, 107203 (2011).
- [30] Y. Song and H. Dery, *Phys. Rev. B* **86**, 085201 (2012).
- [31] K. Sugihara, *J. Phys. Soc. Jpn.* **18**, 961 (1963).
- [32] T. Shimizu, *J. Phys. Soc. Jpn.* **28**, 1468 (1970).
- [33] Y. Yafet, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1963), Vol. 14.
- [34] J. M. Luttinger, *Phys. Rev.* **102**, 1030 (1956).
- [35] G. L. Bir and G. E. Pikus, *Symmetry and strain-induced effects in semiconductors*, (Halsted Press, Jerusalem, 1974).
- [36] R. L. Aggarwal and A. K. Ramdas, *Phys. Rev.* **140**, A1246 (1965).
- [37] P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors* (Springer, Berlin, 2005), 3rd ed.
- [38] We obtain 7.2% true $3d$ weight inside the $1s$ donor envelope. This SOC coupling is estimated to be $0.3 \mu\text{eV}$ [31]. It is partially compensated by the H_{int-d} integral due to larger $3d$ spread. From an upper-limit estimate [39], $J \propto \exp[-d(\frac{1}{a_B} + \frac{1}{3a_B})]$ where a_B is the Bohr radius in Si (before considering a suppressing power-law factor), it is

sufficient to tell that the exchange integral does not completely compensate the 100 times smaller SOC coupling (and another 3.3 times from the energy denominator) for typical $d/a_B \lesssim 5$.

- [39] C. Herring, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press, New York, 1966), Vol. IIB.
- [40] H. Ehrenreich and A. W. Overhauser, Phys. Rev. **104**,

331 (1956).

- [41] The solid angle integration $\int d\Omega_{\mathbf{q}} |\tilde{\epsilon}_{1(2),3,4,LA}|^2 = \frac{4\pi}{15}q^2, \frac{2\pi}{15}q^2, \frac{28\pi}{15}q^2$, and $\int d\Omega_{\mathbf{q}} \sum_{i=1,2} |\tilde{\epsilon}_{1(2),3,4,TA_i}|^2 = \frac{2\pi}{5}q^2, \frac{\pi}{5}q^2, \frac{2\pi}{15}q^2$, respectively.